Metals Recovery: Study of the Kinetic Aspects of Copper Acidic Leaching Waste Printed Circuit Boards from Discarded Mobile Phones

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This study was performed to investigate the possibility of copper recovery from the waste printed circuit boards (WPCBs) of waste mobile phones using a hydrometallurgical route as cleaner technology for environmental protection. In recycling, elements such as copper and aluminium are recovered, as well as the precious metals gold, silver, platinum and palladium, which can be recouped profitably from electronic waste and make this waste very valuable for recycling. In the experiments, the conventional agitated acid leaching method was used for mechanically pre-treated WPCBs with a mean Cu content of 19.09 wt % in a sulphuric acid medium with the presence of Fe^{3+} as an oxidant. The effects of $Fe_2(SO_4)_3$ concentration, leaching time, temperature and the ratio of liquid to solid phase on the recovery of copper from the solutions were studied. The highest Cu extraction was achieved at 363 K after 15 min of leaching, and a liquid to solid ratio (L/S) of 20. The value of activation energy (Ea), which was found to be ~14.87 $kJ.mol^{-1}$, indicates that this process is diffusion controlled. The apparent order of reaction with regard to the initial $Fe_2(SO_4)_3$ concentration at 313 K was calculated as 0.55. The proposed scheme of copper recovery from WPCBs of mobile phones was designed.

Introduction

Nowadays, "waste of electrical and electronic equipment" (WEEE) such as discarded mobile phones (MP), computers or TV sets is one of the world's fastest-growing problems and needs to be tackled immediately. In the European Union (EU) alone, the amount of WEEE is expected to grow to 12 million tonnes by 2020 ("Waste Electrical & Electronic Equipment (WEEE)," 2018). Statistics also indicate that in 2019 the number of mobile phone users will reach 4.68 billion in the world ("Number of mobile phone users worldwide from 2013 to 2019 (in billions)," 2018). WEEE, which consists of very complex materials and components, may cause major environmental and health problems, if not properly managed. Besides, the production of modern electronics needs the use of scarce and expensive resources such as gold, silver and rare earth elements. A mobile phone consists of several parts, including the display unit, battery, front and back cases and printed circuit boards (WPCBs). WPCBs in mobile phones contain many base, precious and rare metals, such as Au, Ag, Pd, Ta, Nd, Al, Cu, Sn, Co, Fe, and Pb. These elements or their alloys are found inside or on the surface of WPCBs. (Palmieri et al., 2014; Sarath et al., 2015). The most attention to the treatment of discarded mobile phones is focused on copper, due to its high content in phones and its current world consumption. The total world production of refined copper was reached 23.4 million tonnes in 2016 (Brown, 2018) ("Secondary copper production up 6% in the first quarter of 2018," 2018). Future global copper demand is expected to keep increasing due to copper's absolutely necessary role in modern technologies. Due to the above-mentioned concerns, these WPCBs have to be removed from discarded mobile phones and selectively treated (European Parliament & The Council Of The European Union, 2012). In general, there are three basic ways to recover valuable metals from secondary resources: pyrometallurgical, hydrometallurgical and a combined method (Moskalyk and Alfantazi, 2003; Behnamfard et al, 2013; Kasper et al., 2018; Rudnik et al., 2015; Birloaga et al., 2013; Havlik et al., 2011). In the EU, the biggest companies use pyrometallurgical treatment methods based on smelting for metal recovery from WEEEs on an industrial scale (Rocchetti et al., 2018). For example, tin, lead, zinc, nickel and precious metals are recovered from electronic scraps, e.g. printed circuit boards, using the Kayser Recycling System (KRS) in Aurubis, Lumen in Germany (Maurell-Lopez et al., 2011; Laubertová et al., 2017). Another example is the Kaldo Furnace process practised at Boliden, Ronnskar in Sweden (Cui and Zhang, 2008) ("Boliden," 2018). The electronic scraps are first smelted in an IsaSmelt furnace to recover precious metals along with Cu in the form of Cu-bullion in Umicore's integrated metal smelter and refinery (Hagelüken, 2006). These smelters can treat more than 200,000 ton/year of precious metal-bearing materials such as by-products from non-ferrous companies and electronic scraps. The hydrometallurgical processes are an alternative to pyrometallurgical treatments of WEEE due to some characteristic advantages such as the possibility of treating heterogeneous materials, lower environmental impact including low gas emission, and

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selective recovery of elements (Laubertova et al., 2011). Some studies exist which focus on treating WPCBs with hydrometallurgical methods (Jing-ying et al., 2012; Zazycki et al., 2017). For example, Kim et al. (2011) leached small pieces of WPCBs (2-3 mm) from discarded MPs in a two-stage leaching process. They selective dissolved copper and gold using electro-generated chlorine as an oxidant. They then leached the crushed WPCBs in an H_2SO_4 acid solution with the addition of H_2O_2 as an oxidant for copper recovery (Camelino et al., 2015; Yang et al., 2011) used combinations of supercritical water and diluted hydrochloric acid leaching methods to recover copper from WPCBs (Xiu and Zhang, 2015). Laubertova et al. (2012) leached the different fractions of WPCBs in either ferric sulfate or a mixture of $FeCl_3$ and $Fe_2(SO_4)_3$ solutions (Laubertova and Sandorova, 2013; Yang et al., 2011). Bioleaching has also been used to leach out gold and copper from waste mobile phone WPCBs (Chi et al., 2011). As seen in the literature, the different fraction of WPCBs of MPs and different acid leaching media were used for the leaching process. A kinetic investigation was also carried out to evaluate the kinetic constants during the leaching processes in different acid leaching media (Dutta at al., 2018; Ha et al., 2014; Kim et al., 2011). According to the literature (Havlik, 2008), a thermodynamic study of the leaching process of copper indicates that it is possible to use an acid oxidizing medium for leaching. However, these studies did not describe the kinetic aspects of leaching of WPCBs in ferric sulphate as a solvent medium. The present research attempts to develop an alternative process for the leaching of copper from mechanically pre-treated WPCBs of MPs using ferric sulphate as a leaching solution. Thermodynamic data were determined using Outotec HSC Chemistry Software from Outotec Research with Oy modelling package 8.0 (Roine, 2002).

The leaching of WPCBs in $Fe_2(SO_4)_3$ solutions is described by chemical reaction Eq. 1:
$$Cu + 2Fe^{3+} + SO_4^{2-} = 2Fe^{2+} + CuSO_4 \qquad \Delta G_{293}^0 = -64.54\,kJ \qquad (1)$$
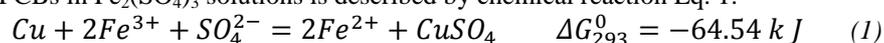
The negative value of the standard Gibbs energy suggests that this reaction is thermodynamically feasible and spontaneous. The objectives of this study are listed as follows:

1) Determination of the chemical composition of Cu-containing wastes (WPCBs from waste button mobile phones).

2) Cu recovery by means of agitated leaching of WPCBs from MPs with different parameters such as liquid to solid ratio, reaction temperature, leaching times and $Fe_2(SO_4)_3$ concentrations.

3) Based on kinetics measurements, obtaining values of the apparent activation energy Ea and the apparent reaction order n, with regard to the initial $Fe_2(SO_4)_3$ concentration in the leaching solution.

Materials and methods

Sample characterization

Discarded MPs collected in a recycling facility in Slovakia were used in these experiments. The sampling procedure of WPCBs shown in Fig. 1 was used for preparing a representative sample for chemical analysis (Laubertova et al., 2018; Mickova, 2018).

Fig. 1. Methodology for printed circuit boards sampling from discarded mobile phones

After manual dismantling and sorting of MPs, the non-magnetic fraction of WPCBs was mechanically milled and then sieved to less than 1.25 mm. In order to evaluate the size distribution of the treated sample, 100 g of the selected sample obtained by quartering was sieved with a vibration sieve shaker using 1.25; 0.71; 0.65;

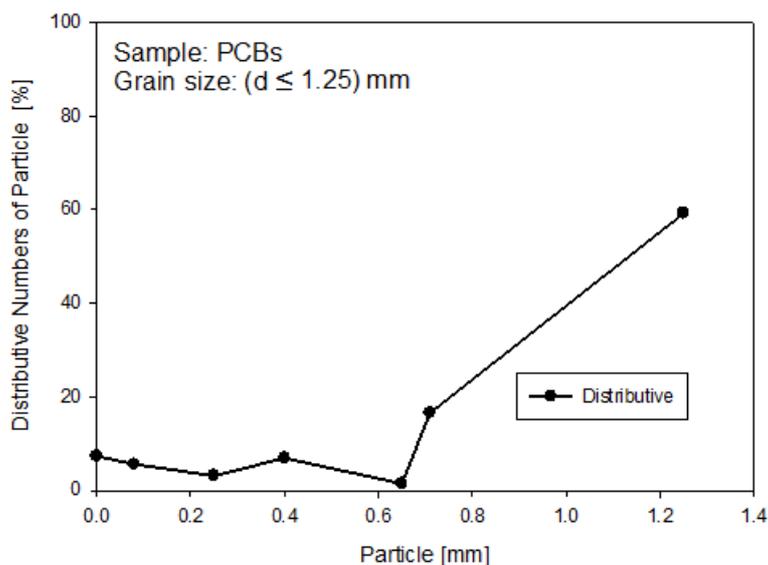0.40; 0.25; 0.08 mm openings. The size distribution of WPCB samples used for the experiments is shown in Fig.2.

Fig. 2. Particle distribution of the treated sample WPCBs (d ≤ 1.25 mm)

X-ray diffractometer Phillips PW 1710 X´Pert PRO MRD (Co-Kα) was used for qualitative phase analysis. Sample evaluation was performed using software RIFRAN. Due to the fact that it is not possible to obtain an objective diffractogram from such large particles, the samples were milled on a vibratory mill to obtain a fine-grained homogeneous fraction suitable for X-ray diffraction qualitative analysis. Diffraction pattern of the sample is shown in Fig. 3. The results of the phase analysis show that all samples contain metals and / or Cu, Sn, Pb, Zn, Fe alloys. The proportion of plastics represented by the high background of the diffractogram in the region around 25 ° Bragg angle 2Theta is significantly represented. This fact makes the identification more difficult.
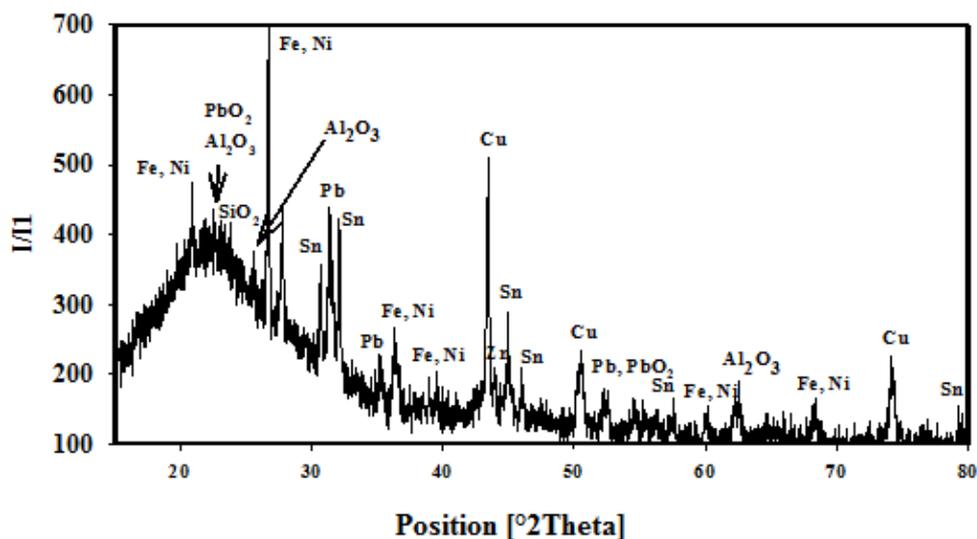

Fig. 3. XRD pattern of the fine-grained fraction of the WPCBs

Five representative samples (1g each) taken from treated WPCBs (d ≤ 1.25 mm) were subjected to atomic absorption spectroscopy (AAS) (Varian Spectrometer AA 20+) analysis to determine the concentrations of the selected metals. The average results of the elemental analysis of the WPCB samples are shown in Table 1.

Table.1. Average results of elemental analysis of five representative WPCBs samples using the AAS technique

Element	Cu	Ni	Sn	Zn	Pb	Al	Fe	Au	Ag
Content (wt %)	19.09	1.26	1.286	1.776	0.403	0.826	1.956	0.036	0.093
Standard deviation „s"	6.304	0.341	0.629	1.21	0.202	0.249	1.405	0.005	0.021
Coefficient of variation „C$_V$"	0.33	0.26	0.48	0.68	0.5	0.30	0.718	0.157	0.223

Experimental procedure

$Fe_2(SO_4)_3$ and H_2SO_4 were used for leaching representative WPCB samples in an aqueous medium. The leaching experiments were conducted in a conventional reactor with a stirrer at 400 RPM. The effect the concentration of the solution on the leaching rate was investigated in the concentration range from 0.1 mol dm^{-3} to 1 mol dm^{-3} $Fe_2(SO_4)_3$ at 0.5 mol dm^{-3} H_2SO_4. The used liquid to solid (L/S) ratios were 10, 20 and 30 (200 mL solution/20-10-6.6 g solids). The effect of temperature on the leaching rate was also investigated at temperatures between 314 K and 363 K in the solution containing 1 $Fe_2(SO_4)_3$+0.5 mol dm^{-3} H_2SO_4. The schematic representation of the experimental set-up is shown in Fig. 4. A solution mixture of sulphuric acid and Fe^{3+} as an oxidizing agent were used for leaching. For all experiments, 0.5 mol dm^{-3} H_2SO_4 was oxidized with 1 mol dm^{-3} $Fe_2(SO_4)_3$. The leaching time for each experiment was 60 minutes. Every 5, 10, 15, 30 and 60 minutes a 5 ml sample was taken for chemical analysis. The samples were subjected to atomic absorption spectroscopy for determination of Cu content.
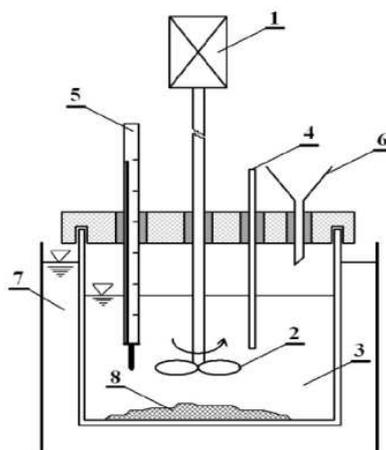

Fig. 4. Schematic diagram of the conventional agitated leaching system (Trung et al., 2011)
1- a drive of the mixer with constantly adjustable stirring, 2 – mixer, 3 – leaching agent,
4 – liquid sample collection, 5 – thermometer, 6 – material input, 7- thermostat, 8 – sample

Results and discussion

Before the experimental investigation, a computational thermochemical study was carried out to predict the possible phases of the system depending on changing parameters. The classical Eh-pH (Pourbaix) diagrams are known for being similar to "predominance diagrams" which show the regions where various aqueous ions or solid compounds predominate (Bale et al., 2016). In this study, a new type of aqueous phase diagram named FactSage 7.2 was used to separate the aqueous species from the solid phases by real phase boundaries. The diagram was calculated at constant molalities of Cu, Fe, Zn, and Ni as used in the experiments. In the calculations, the Pitzer database for concentrated solutions was used, and the temperature and total pressure of the system were selected as 80 °C and 1 atm respectively. The aqueous phase diagram of our H_2O-$Fe_2(SO_4)_3$-H_2SO_4-Cu-Zn-Ni system calculated with the FactSage phase diagram module is shown in Fig. 5. The y-axis shows the oxidation potential, log P(O_2), which is related to Eh, while the x-axis is the $Fe_2(SO_4)_3$ concentration

given as the molality (mol/kg H_2O) in the solution. As seen in the figure, when the molality of H_2SO_4 was fixed at 0.5 in the system, there was a stable aqueous region throughout the system for a certain oxygen partial pressure range (i.e. $\log_{10}PO_2$, between -35 and -50 atm). When the oxygen amount increased in the system, Fe_2O_3 precipitated out from the solution. In reducing conditions, in addition to gas, solid Cu, ferrous sulphate, and iron oxides started precipitating out. For example, when the oxidation potential and the molality of $Fe_2(SO_4)_3$ were selected as -40 atm and 1 respectively, all metallic elements (i.e. Cu, Zn, Ni, and Fe) in the aqueous solution were calculated to be almost entirely in the (2+) oxidation states.
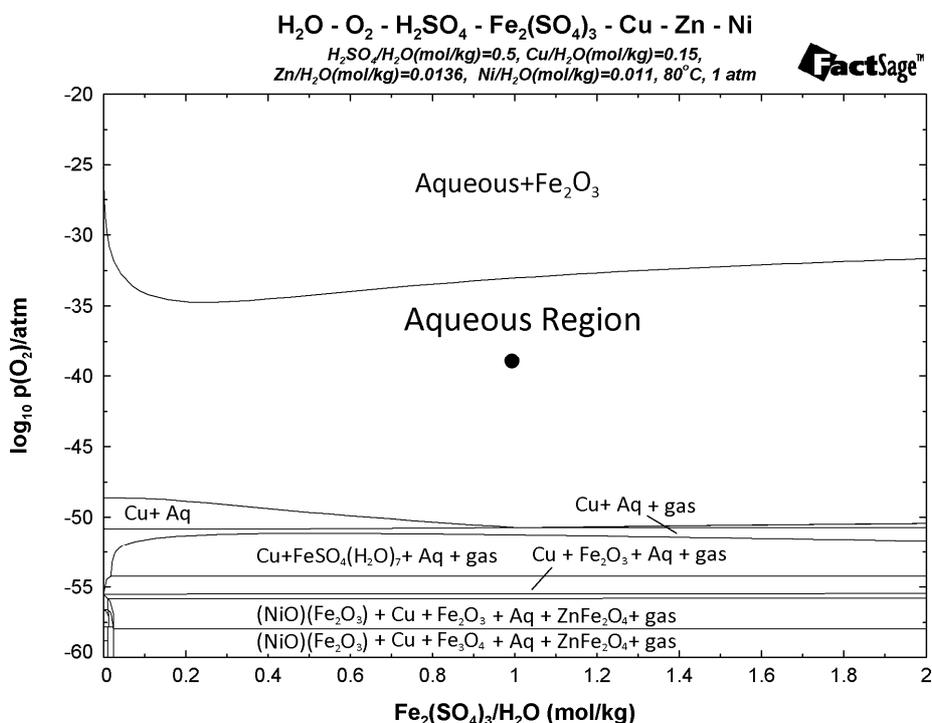

Fig. 5. Aqueous phase diagram – calculated partial pressure of O_2 vs molality of $Fe_2(SO_4)_3$ for the system H_2O-$Fe_2(SO_4)_3$-H_2SO_4-Cu-Zn-Ni under one of the present experimental conditions

Effect of liquid to solid phase ratio on the leaching

The weights of 20, 10 and 6.6 g of samples were leached in 200 ml of a solution containing 0.5 mol dm^{-3} $Fe_2(SO_4)_3$ + 0.5 mol dm^{-3} H_2SO_4 to determine the copper extraction yield from the PCB samples.   Fig. 6 shows that the highest extraction (47.95 %) was achieved when the experiment was performed at L:S ratio of 20, at 333 K, with stirring rate 6.6 s^{-1} and 10 minutes of leaching.
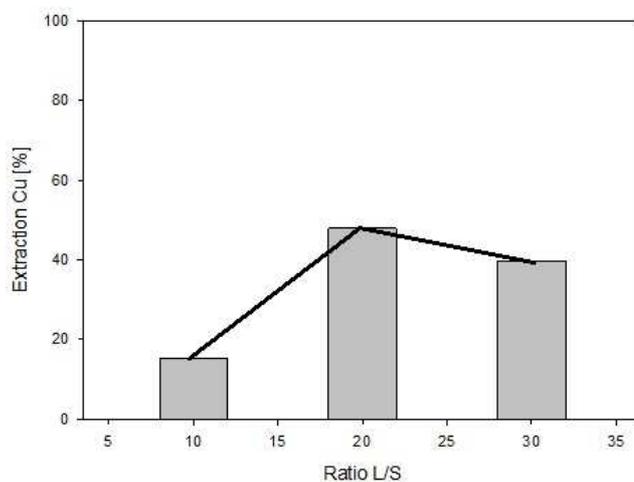

Fig. 6. Effect of liquid to solid ratio on copper extraction with 10 min. leaching

Effect of temperature

The leaching experiments were carried out at 313 K, 333 K, 353 K and 363 K and a liquid to solid ratio L/S 20 for the solution of 1 mol dm^{-3} Fe$_2$(SO$_4$)$_3$ + 0.5 mol dm^{-3} H$_2$SO$_4$ after 60 min of leaching. The positive effect on copper extraction of increasing leaching temperature is shown in Fig. 7. The highest Cu extraction (69.98 %) was achieved at 363 K after 15 min of leaching. No significant change was observed in Cu extraction after 30 minutes.
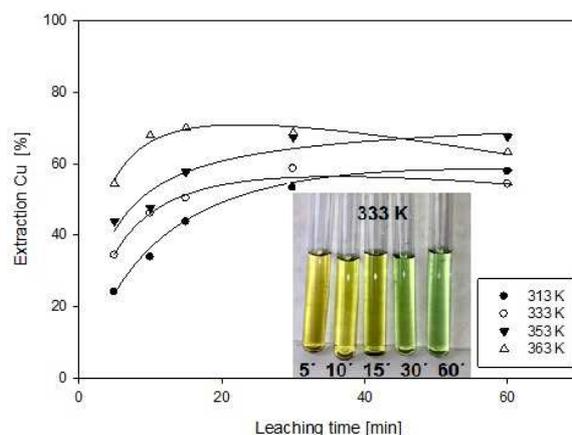

Fig. 7. Effect of temperature on copper extraction with time (1 mol dm^{-3}Fe$_2$(SO$_4$)$_3$ + 0.5 mol.dm^{-3} H$_2$SO$_4$, L/S 20, stirring rate 6.6 s^{-1})

The particle morphology of the treated sample, determined with a digital microscope (Dino-Lite ProAM413T), showed that the metallic parts were heterogeneously distributed throughout the sample (Fig.8a.). The sample was also studied with a digital microscope before and after leaching at 353 K and 60 min. The following figures show the morphological structures of the individual sample upon visual observation. It can be seen in Fig. 8 (a-b) that most of the metallic parts **X** found in the sample Fig.8a were dissolved after leaching sample **Y** in Fig. 9b.
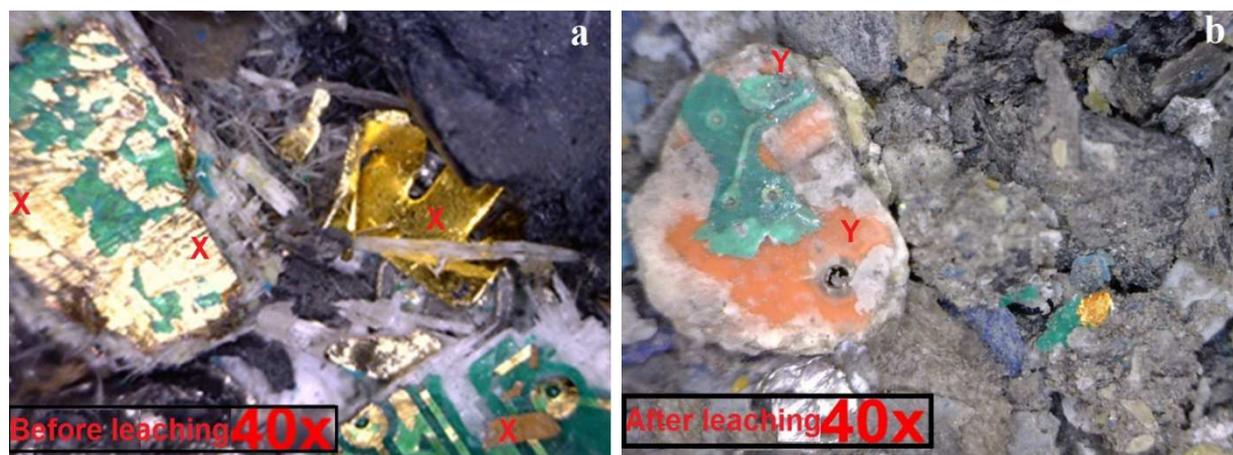

Fig. 8. Samples printed circuit boards a) before leaching, and b) after leaching at 353 K and 60 min (1 mol dm^{-3} Fe$_2$(SO$_4$)$_3$ + 0.5 mol.dm^{-3} H$_2$SO$_4$, L/S 10)

Effect of Fe$_2$(SO$_4$)$_3$ concentration

Fig. 9 shows the effect of leaching time on copper recovery with changing ferric sulfate concentrations ranging from 0.1 to 1 mol dm^{-3} Fe$_2$(SO$_4$)$_3$ at 0.5 mol dm^{-3} H$_2$SO$_4$ with L:S ratio of 20 at 313 K. The copper yield significantly increased with increasing concentration of ferric sulphate. The highest extraction of Cu (58.73 %) was achieved at 313 K after 30 min of leaching. Longer leaching durations did not give any more satisfactory result beyond 30 min.
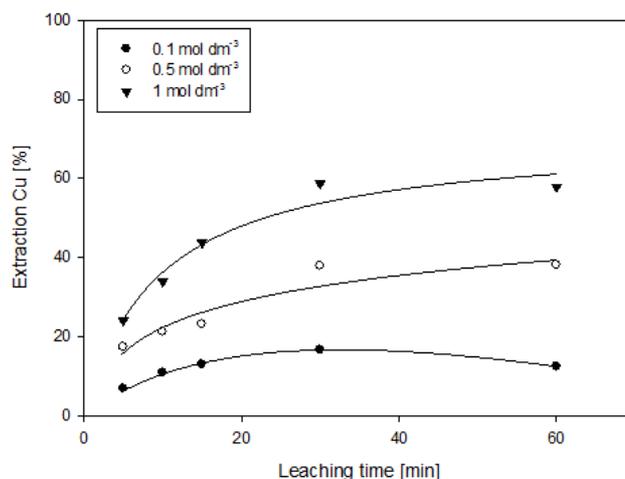
Fig. 9. Effect of $Fe_2(SO_4)_3$ concentration on copper extraction L/S 20, 6.6 s^{-1} and 313 K

Apparent Activation Energy "Ea" and Apparent Order of Reaction "n"

The apparent activation energy Ea was determined in the temperature interval of 313-363 K for the leaching solution of 1 mol.dm^{-3} $Fe_2(SO_4)_3$ + 0.5H_2SO_4 mol dm^{-3}. The apparent activation energy **Ea** was calculated experimentally by measuring the initial dissolution rate of copper at different temperatures **T** for the time interval of 0-300 s. The Arrhenius equation was used to determine the activation energy Eq. 2:

$$ln\, k = lnA - E_a/R\,.T \qquad (2)$$

where **k** is the rate constant, **A** is the frequency factor, **R** is the universal gas constant, and T is the temperature. The estimated **Ea** apparent activation energy for the sample was found to be 14.87 kJ mol^{-1} in the temperature interval from 313K to 363 K, and the frequency factor **A** was calculated as 0.2427 s^{-1}. It is generally believed that if the value of activation energy is higher than 40 kJ/mol, the process is chemically controlled, whereas values less than 20 kJ/mol suggest that the process is controlled by diffusion (Habashi, 1997).

The estimated Ea (14.87 kJ mol^{-1}) indicates that the leaching process is controlled by diffusion. The corresponding relationship between **ln k** and **1000/T** is shown in Fig. 10, which indicates that the mechanism for sample leaching in the temperature interval 313-363K does not change.

The apparent activation energy **E$_a$** and the apparent order of reaction **n** were determined using the linear regression method from Eq. 2 and Eq. 3, respectively (Cao et al., 2006; Kim and Lee, 2016).

The following concentration interval was used for determining the apparent order of reaction at temperature 313 K: from 0.1mol dm^{-3} to 1 mol dm^{-3} $Fe_2(SO_4)_3$.

The apparent order of reaction **n** with regard to the initial concentration of $Fe_2(SO_4)_3$ in the leaching solution was determined according to the following Eq. 3:

$$v_{Cu} = k.\,c_{Fe_2} \qquad (3)$$

or in logarithmic form Eq. 4:

$$lnv = lnk + n.\,lnc_{Fe_2} \qquad (4)$$

where **n** is the slope of the graph of $lnv_{Cu} = f\,c_{Fe_2}$, shown in Fig. 11. The value of $n = 0.55$ and $ln\,k = $ -7.104. The dissolution of Cu at the temperature of 313 K can be represented by the following Eq.5:

$$v = 8.21.\,10^{-4}c_{Fe_2} \qquad (5)$$

or in logarithmic form Eq. 6:

$$lnv = -7.104 + 0.55.lnc_{Fe_2} \qquad (6)$$

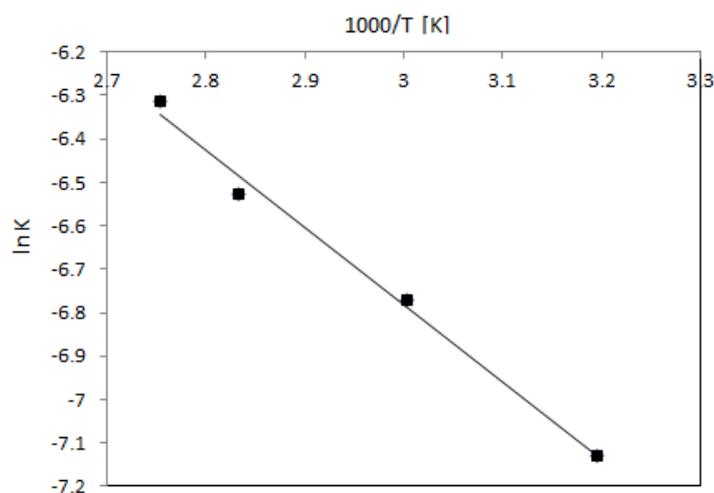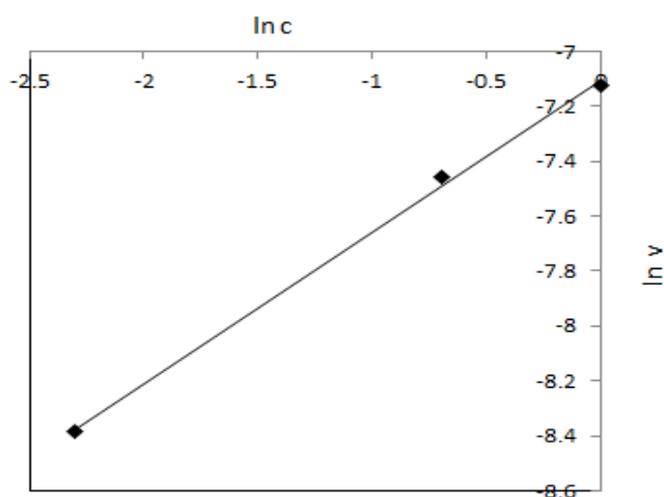*Fig. 10. Arrhenius plot for copper extraction from the waste sample: correlation coefficient R = -0.9957*

Fig 11. Logarithmic plot ln v versus ln c_{Fe_2} for waste sample: Correlation coefficient R = 0.99904

The result of the investigation was the finding that the best copper yields were achieved using 1 mol dm^{-3} Fe$_2$(SO$_4$)$_3$ + 0.5mol dm^{-3} H$_2$SO$_4$, at a temperature of 333K and liquid to solid phase ratio L:S 20. The conclusion and the ensuing recommendation was that the effect of milled mobile phones on Cu should be determined.

A cementation treatment has been proposed for the further treatment of the leached solution to precipitate out the copper as a saleable product in the presence of solid iron finings. The results of these experiments have been published elsewhere (Kovalcik, 2017). The proposed process flow sheet for the recovery of Cu from waste mobile phones is shown in Fig. 12
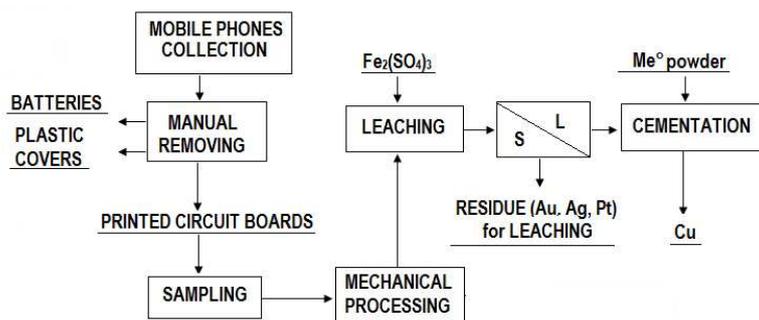
Fig.12. The proposed scheme of copper recovery from waste PCBs of mobile phones

Conclusions

Waste printed circuit boards from discarded mobile phones contain many interesting raw materials - base (copper 15%), precious (gold 400 g t), and rare metals. In the European Union, the biggest companies use pyrometallurgical process treatment methods based on smelting for metal recovery from electronic scrap on an industrial scale. The hydrometallurgy process of treatment printed circuit boards can achieve significantly lower environmental impacts than pyrometallurgy. From a life cycle perspective, the introduction of new technologies can also contribute to reducing greenhouse gases and, possibly, other environmental impacts.

The effects of temperature and concentration of ferric sulphate solution on the leaching rate of copper from milled discarded PCBs from button mobile phones were studied using the agitated leaching process in this research. The results show that the temperature has a significant effect on the leading rate of copper. According to the Arrhenius equation, the apparent activation energy value of the copper reaction for apparent activation energy value of copper reaction for the sample is calculated to be 14.87 kJ mol, under these conditions. On the basis of this value, it may be suggested that the process of leaching takes place in the diffusion region. The highest extraction Cu yield (69.98 %) was achieved at 363 K after 15 min of leaching when an L/S ratio of 20 and a solution of 1 mol dm^{-3} $Fe_2(SO_4)_3$ + 0.5 mol dm^{-3} were used. The apparent order of reaction with regard to the initial $Fe_2(SO_4)_3$ concentration was n = 0.55. Waste electronic materials such as discarded mobile phones are a secondary source of critical materials for the European Union. Hydrometallurgy could be one of the possible innovative ways to metals recovery from printed circuit boards of electronic waste materials.

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